

## Survey and Exposure Estimates of Uranium in Desert Lettuce

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### Introduction

Food safety issues are of growing concern to consumers. Both microbial and inorganic contaminants are of concern in fresh vegetables. This concern over inorganic contaminants is exemplified by recent publicity over the small levels of perchlorate in the Colorado River and accumulation by crops irrigated with this water. Recently, we conducted survey work for perchlorate in lettuce and we found potential perchlorate exposure from lettuce was negligible relative to harmful amounts. By developing this information we successfully avoided a food safety scare that might have had an adverse economic impact on local produce industries.

More recently there has been publicity concerning uranium in the Colorado River (American Rivers, 2004). Uranium (U) is a health concern as a potential carcinogen and as a causal agent of kidney dysfunction (Yuile, 1973; Stevens et al., 1980; Morrow et al., 1982). Human exposure to U can be through dermal contact, inhalation, or ingestion through water or food. In natural environments, uranium isotopes include  $^{238}\text{U}$  (99.3%),  $^{235}\text{U}$  (0.71%), and  $^{234}\text{U}$  (0.006%). Natural uranium is found in granites, metamorphic rocks, lignite, monazite sands, phosphate deposits and other geological materials, including soils (Cothren and Lappenbusch, 1983; Zhang et al. 2002). For the water the samples we collected we observed ratios consistent with natural U in equilibrium.

Based on a national survey it was reported that the intake of uranium in a typical diet is 1.75 ug/day (Welford and Baird, 1967). The National Council on Radiation Protection and Measurements (NCRPM) has estimated that humans take in approximately the same amount of uranium in food as they do in drinking water (NCRPM, 1984). The 1999 guidance from EPA on gastronomic absorption of uranium gives gastronomic absorption fractions as 4 percent for infants, 2 percent for children between 1 and 15 years of age, and 2 percent for adults (USEPA, 1999).

The kidney is a recognized target organ for uranium toxicity. Effects have been observed in both studies with laboratory animals (Gilman et al., 1998a; 1998b; 1998c) and humans (Zamora *et al.*, 1988). Wilkinson (1986) reports an increased rate of mortality from gastric cancer in counties of northern New Mexico having significant deposits of uranium compared to counties without significant uranium deposits. In another study, Health Canada (1998) found there was a positive correlation between oral exposure to uranium and tubule effects in kidneys. These data suggest an excretion 'No observed effect level' (NOEL) of 0.12 ug/day. Assuming a gastronomic absorption of 2% the NOEL for uranium intake would be approximately 6 µg/day.

The U.S. Environmental Protection Agency has established a maximum contaminant level (MCL) for natural uranium of 30 µg /L (U.S. EPA, 2000). The state of California has an MCL for uranium of 20 pCi/L based on earlier studies of toxicity to the kidneys in rabbits. But based on more recent clinical and epidemiological evidence the State of California has set the Public Health Goal (PHG) for uranium at 0.5 µg/L (OHHEA, 2001).

We have no information on the source of uranium in the Lower Colorado River. It has been alleged that one significant source may be the abandoned Atlas mill site near Moab, Utah (American Rivers, 2004). This former uranium ore processing facility accumulated 10.5 million tons of uranium mill tailings during its operation until 1984 (DOE, 2004). This Uranium Mill Tailing Remedial Action (UMTRA) site has been designated a Title II site in that the mill tailing remain in place and it potentially contributes high levels of U to aquifers near the source. This particular site has among the largest plumes as measured by concentrations of 10 to 20 pCi/L in sampling wells (Jove-Colon et al., 2001). It has been alleged that approximately 110,000 gallons of radioactive waste seep into the Colorado River each day from an unlined impoundment where 12 million gallons of radioactive waste is stored (American Rivers, 2004). The Department of Energy has implemented a plan at stabilization of this site (DOE, 2004). While there is an urgent need to stabilize the UMPTRA site near Moab, it is unlikely the only significant source of U to the Colorado River. Owing to geology of the region and a history of mining we would also anticipate contributions from tributaries north and south of the Moab site as well (southern Colorado and Utah and Arizona).

More recently, we have measured uranium concentrations at the Imperial Diversion Dam near Yuma, Arizona of 3 to 5 µg /L. These ranges are not excessive considering ranges found in many western water sources (Wong et al., 1999). However, the lower Colorado is used extensively for irrigation of food crops shipped throughout North America and the potential for enhanced human exposure nationally through food is of potential concern. The objective of this study is to demonstrate that the levels of U in lettuce pose a negligible health risk.

## **Materials and Methods**

### **Uptake of Uranium by lettuce as affected by content in Colorado River Water**

A greenhouse experiment was conducted to evaluate uptake of uranium from Colorado River water. In these experiments we used soils of varying cropping history and silica sand. For this study we used 1.5 L pots with an outside diameter of 15 cm each filled with approximately 1.5 kg of soil or sand. The experiment consisted of two water treatments, irrigation with Colorado River water containing natural concentrations of uranium and synthetic Colorado River water free of uranium. The experimental design was completely random with 4 replications. Lettuce was seeded in trays initially and subsequently transplanted into the experimental pots. This experiment was harvested

after 60 days and above ground plant material and roots will be weighed, oven-dried, and ground for analysis. Uranium was determined after digestion using ICP/MS.

### **Survey and Exposure of Uranium in Lettuce**

Lettuce samples were collected in the lower Colorado River Valley of Arizona. Crop samples brought into the laboratory were diced, mixed thoroughly, and a sub-sample placed in the freezer. Samples were freeze-dried, and ground, and digested using nitric acid and peroxide. Uranium was determined by ICP/MS. Uranium levels in lettuce will be used to form human exposure assessments. We used data prepared from U. S. food consumption surveys (Smiciklas-Wright, 2002) for these exposure estimates.

### **Results and Discussion**

We have found the total uranium in the Colorado River to range from 3 to 5  $\mu\text{g/L}$  at the Imperial Diversion Dam. As expected for natural uranium, over 99% is the U238 isotope. However, very low levels of U235 are detectable using standard ICP/MS methodology. Samples we collected along the entire channel of the Colorado River and selected tributaries show that the near head waters, the content of uranium in the river is barely detectable. However, the Colorado River begins to take on higher concentrations as the river descends onto the Colorado Plateau (Table 1). Concentrations of total uranium within the main channel range from 3 to 5  $\mu\text{g/L}$  from Grand Junction, Colorado to Yuma, Arizona. However, the river is not the only source of uranium to cropping systems in the region. We have found the uranium contents of mono-ammonium phosphate commonly used for lettuce production in the region to average xxxxx mg/kg

The results of the greenhouse study show no difference in uranium uptake by lettuce to natural Colorado River water containing uranium and synthetic Colorado River water containing no uranium (Table 2). These data suggest water is not the most significant source of uranium during the growing season. This does not preclude the possibility that over time, uranium added with water accumulates in the soil where it affects uptake over time.

There are significant differences in uranium uptake to soil (Table 2). This may reflect differences in the natural uranium contents of the soils, it may reflect different phosphorus fertilization histories, or it may be due to the fact that certain soils are more inclined to retain the uranium added with water and phosphorus fertilizers.

The concentrations of total uranium in lettuce produced in the fields of the lower Colorado River region range from 0.1 to 13.5  $\mu\text{g/kg fw}$  (Table 3). Concentrations were generally higher in the harvested portion of leaf lettuce types as compared to iceberg. Estimated adult male dosages ranged from 0.0002 to 0.003  $\mu\text{g/kg bw-day}$ . The USEPA has established a reference (RfD) dose of 3  $\mu\text{g/kg bw-day}$  for uranium. Generally, uranium intake from lettuce would be less than 0.1% the RfD.

In summary, the Colorado River contains low levels of uranium and phosphate fertilizers used for lettuce production in the region also contain uranium. Lettuce produced in the region also accumulates trace amounts of uranium. However, the levels of uranium consumed with lettuce are very low relative the EPA RfD, a dose generally considered safe.

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Table 1. Summary of selected uranium found in water samples collected at several locations along the Colorado River or it's tributaries.

River	Description	U <sup>238</sup> (µg/L)	U <sup>235</sup> (ng/L)
Colorado	Grand Lake, Co	0.1	0.1
Colorado	Kremmling, Co	2.0	10.3
Colorado	Glenwood Springs, Co	1.5	8.0
Colorado	De Bisque, Co	1.6	8.1
Gunnison	Grand Junction, Co	7.6	39.3
Dolores	Gateway, Co	2.8	13.3
Colorado	Upstream of Moab, UT	5.3	27.0
Colorado	Downstream of Moab UT	6.1	27.7
Green	Green River, UT	2.8	14.8
San Rafael	SW of Green River, UT	6.7	33.6
Dirty Devil	W of Hanksville, UT	4.3	22.8
Colorado	Hite, UT	5.2	26.6
San Juan	Mexican Hat, UT	2.8	14.2
Colorado	Page, AZ	3.7	20.5
Paria	Lees Ferry, AZ	10.7	54.9
Colorado	Lees Ferry, AZ	3.6	19.4
Little Colorado	Cameron, AZ	7.4	43.0
Virgin	SW of Mesquite, NV	5.7	28.4
Muddy	Overton, NV	13.3	69.8
Las Vegas Wash	Lake Mead	6.5	32.7
Colorado	Willow	4.5	26.2
Colorado	Lake Havasu City	5.2	24.6
Bill Williams	S of Lake Havasu City	4.7	24.3
Colorado	Below Parker Dam	5.2	23.6
Colorado	Imperial Diversion	4.3	21.5
Gila	N of Yuma	6.9	37.2
Colorado	Morelos Diversion	4.5	23.6

Table 2. Uranium uptake of lettuce as affected by irrigation water and soil.

<b>Irrigation Water</b>	<b>Uranium (ng/pot)</b>
Synthetic	108
Natural	106
Stat.	NS
<b>Soils</b>	
Silica sand	77
Superstition sand (Desert)	64
Superstition sand (cropped)	164
Indio Silty Clay loam	147
LSD	61

Table 3. Uranium contents of field grown lettuce irrigated with Colorado River water.

Lettuce Type	Isotope	Uranium (µg /kg fw)			
		Min.	Max.	Mean	Median
Iceberg	<sup>235</sup> U	<DL	0.021	0.004	0.004
	<sup>238</sup> U	0.121	1.873	0.386	0.275
Romaine	<sup>235</sup> U	<DL	0.033	0.008	0.005
	<sup>238</sup> U	0.234	2.260	1.087	1.031
Boston	<sup>235</sup> U	0.003	0.037	0.021	0.025
	<sup>238</sup> U	0.588	7.559	3.950	4.072
Green leaf	<sup>235</sup> U	0.001	0.058	0.029	0.214
	<sup>238</sup> U	0.896	10.999	4.154	3.593
Red leaf	<sup>235</sup> U	0.003	0.062	0.024	0.023
	<sup>238</sup> U	1.496	13.494	5.035	4.583

Table 4. Hypothetical uranium exposure from field grown lettuce irrigated with Colorado River water.

Lettuce Type	Mean µg /kg fw	Adult male exposure (µg /day)	Adult male dosage (µg /kg bw-day)
Iceberg	0.386	0.0127 (0.0377)	0.0002 (0.0005)
Romaine	1.087	0.0474 (0.1412)	0.0007 (0.0020)
Boston	4.154	0.1653 (0.4922)	0.0024 (0.0070)
Green leaf	5.035	0.2108 (0.6278)	0.0030 (0.0090)
Red leaf	3.950	0.1873 (0.5578)	0.0027 (0.0080)

Based on mean and 90<sup>th</sup> percentile lettuce consumption of 46 and 137g.  
 Values in parenthesis represent 90<sup>th</sup> percentile values.